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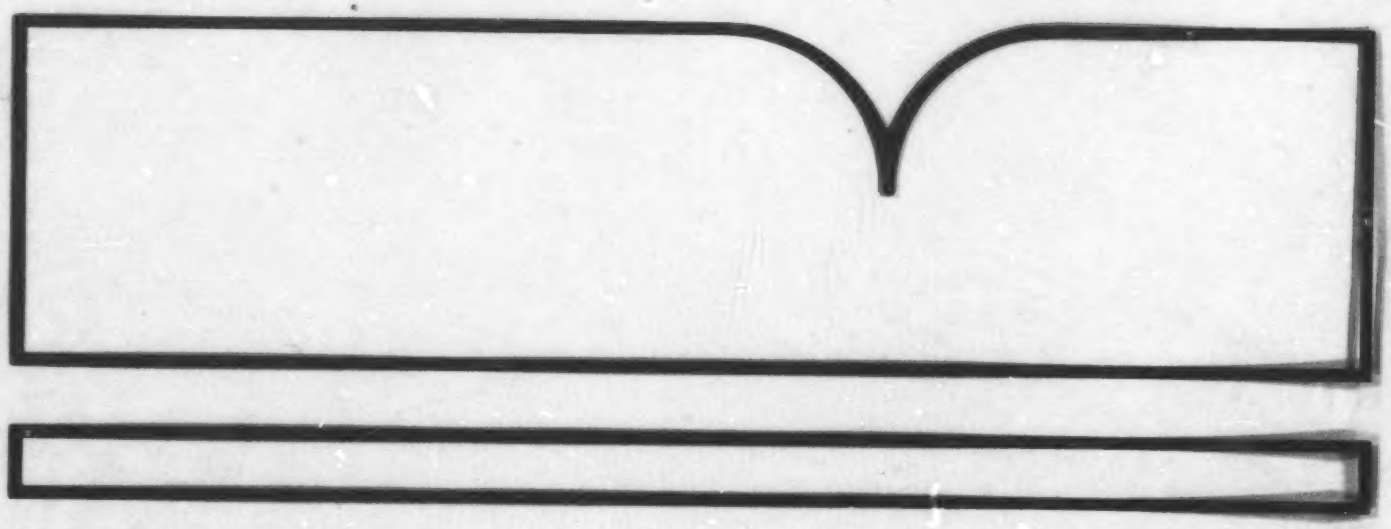

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16. ABSTRACT

Published literature on the environmental fate of pollutants published during 1984 are reviewed. Short exceprts are presented from each reference covering such areas as photolysis, biodegradation, hydrolysis, sorption, and volatility for pollutants including pesticides, hydrocarbons, heavy metals, polynuclear hydrocarbons, and other toxic organic chemicals.

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Fate of environmental pollutants

P. H. Pritchard

XENOBIOTIC CHEMICALS

Several papers described the fate and distribution of polychlorinated biphenyls (PCBs) in aquatic ecosystems. Concentrations of PCBs detected in water, sediments, and biota from the Mediterranean Sea,[1] lakes[2–4], and estuarine areas[3] have been reported. Larsson's[2] observation of higher PCB concentrations in sedimenting particulates than in bed sediments led him to suggest that PCBs deposited in sediment may be mobile and possibly recycled back into the water column. A microcosm system containing field-contaminated sediment was used by Fisher *et al.*[4] to verify that PCBs are released from sediments to overlying water. Release rates depended on chlorine substitutions in the PCB molecule, chemical concentration, and diffusion rates. Sedimentation rates of particulates, however, would slowly eliminate communication of the PCBs in the undisturbed contaminated sediment layer by isolating them from the overlying water. The fate, in microcosms, of PCBs initially adsorbed to clay particles was described by Millard *et al.*[5] as controlled by transfer from the clay particles to particulate organic carbon and by volatilization. Baxter and Sutherland[6] showed that both photolysis and biodegradation contribute to the complete decomposition of 2,4'-dichlorobiphenyl, a combination they suggest could be important in the overall fate of PCB's in aquatic systems. Studies by Massé *et al.*[7] on the biodegradation of 4-chlorobiphenyl by *Achromobacter* sp. and *Bacillus* sp. provided evidence for both a dioxygenase and monoxygenase attack of the aromatic ring in natural systems.

Partial biodegradation of TCDD was reported by Quensen and Matsumura.[8] They showed that solvents (ethyl acetate, DMSO, ethanol) accelerated what seemed to be a hydroxylation of one or both of the benzene rings. Evidence for the chemical alteration of hexachlorocyclopentadiene in water that was used to grow goldfish has also been reported.[9] Field studies in an Argentine estuary[10] described the major fate of hexachlorocyclohexane to be sorption to particulates, followed by precipitation of the contaminated particulates in the freshwater-seawater mixing zone. Rippen *et al.*,[11] using a modeling approach based on production data and use patterns, were unable to reasonably predict environmental concentrations of p-dichlorobenzene and trichloroethene in river water without considering direct inputs of the substances into water. Other studies with chlorobenzenes further characterized possible environmental fates caused by photolytic[12] and biodegradation[13] processes. Jansson *et al.*[14] described an analytical method for determining concentrations of polychlorinated naphthalenes in various types of environmental samples.

The fate of chlorophenols in aquatic ecosystems continued to be a focal point for studies of environmental contamination.[15] photochemistry,[16] sorption to sediments,[17–18] biodegradation,[19–23] and field or microcosm studies.[24–26] Schellenberg *et al.*,[17] for example, characterized chlorophenols as weak hydrophobic acids that sorb to sediments in natural systems by partitioning of the nondissociated species. However, for some of the chlorinated phenols (for example, tetra- and pentachlorophenol), phenolate sorption cannot be ignored, because at ambient pH's, these compounds would be predominately in the ionized form. Biodegradation of chlorophenols was described by Banerjee *et al.*[19] as dependent, in general, on the rate at which the chemical penetrates the bacterial cell membrane and, consequently, on the hydrophobicity of the chemical. Degradation rates of the chlorophenols were inversely related to the degree of chlorine substitution, but unrelated to the site of substitution.

Anaerobic biodegradation of chlorophenols with different degrees of chlorine substitution were also reported.[20,21] In each case, long acclimation times in anaerobic wastewater sludge digesters were required. Edgehill and Finn[22] have successfully decontaminated soil containing high concentrations of pentachlorophenol (PCP) by inoculation with a pure culture of

Arthrobacter, a bacterium that can use PCP aerobically as a sole source of carbon and energy. Their work suggested the possibility of controlling the fate of environmental pollutants by favorable manipulation of the environment. Studies on the fate of chlorophenols in microcosms revealed several important controlling factors that might not be apparent in tests with simpler systems. Inorganic nutrient limitations of microbial activities, adaptation to faster biodegradation rates, and photolysis were the major processes affecting the fate of mono- and dichlorophenol in marine plankton communities contained in large outdoor plastic enclosures.[24] Rooted macrophytes enhanced the biodegradation of pentachlorophenol in littoral microcosms[25] and sorption to sediment particulates, diffusion into sediment beds, and photolysis were the predominant processes that affected the fate of chlorophenols, chlorobiphenyl, and hexachlorobenzene in outdoor experimental ponds.[26]

Aromatic amines and other nitrogen-containing pollutants were shown to degrade in aquatic systems by various complex processes, including biodegradation.[27-33] photolysis,[34] and chemical oxidation.[35] Aniline biodegradation in pond water involved oxidative deamination to catechol and eventual metabolism to CO_2.[27] Trichloroaniline degradation in river water was enhanced by the addition of sediments.[28] Yoneyama and Matsumura[29] demonstrated that 4,4'-methylene-bis-(2-chloroaniline) could be partially biodegraded by pure cultures of bacteria through N-acetylation and N-hydroxylation reactions. The authors suggested, by analogy with mammalian detoxification pathways, that the microbial metabolic products may be mutagenic. Dichloraniline, however, was persistent in laboratory studies and field-enclosure experiments containing natural seawater.[36] Nyholm *et al.*[30] discussed the importance of lag periods for biodegradation in assessing the environmental persistence of *p*-nitrophenol. Use of acclimated inocula was criticial for proper test interpretation. Propanediol dinitrate, while not readily biodegradable, was described by Wyman *et al.*[34] as photolabile and, under certain azeotropic conditions, significantly volatile. Dinitrotoluene[31] and aminobenzoate[32] have been shown to be readily biotransformed under anaerobic conditions, using activated sludge and a pure bacterial culture respectively. The environmental fate of quaternary ammonium compounds was reviewed by Boethling.[33]

The fate of diethylphthalate in microcosm models of freshwater streams was examined by Lewis *et al.*,[37] and was noted to be completely controlled by microbial activities, particularly those associated with Aufwuchs communities. Similar dominance of biodegradation of dibutylphthalate in estuarine systems, particularly when sediments were included in the test systems, was also reported by Walker *et al.*[38] Lag periods before the onset of degradation and first-order decay constants varied widely over six different sampling sites. Johnson *et al.*[39] determined experimentally that complexity of the di-ester alkyl group of phthalic acid esters was the most important factor in determining biodegradability in aquatic systems. Biodegradation was most active in nutrient-rich aquatic systems above 22°C. Previous exposure to phthalate esters, contrary to the results of Walker *et al.*,[38] did not influence biodegradability. Mineralization of various phthalate esters in anaerobic wastewater digesters was also reported.[40] Complexity of the di-ester alkyl group again determined which phthalates might pass, undegraded, through a wastewater treatment system and potentially contaminate receiving waters.

Isopropylphenyl diphenyl phosphate, a triaryl phosphate used as a flame retardant, was shown by Heitkamp *et al.*[41] to be slowly biodegraded in freshwater sediments, despite attempts to vary oxygen tension, chemical concentration, and sampling time. Degradation seemed to proceed through a stepwise demethylation of the substituted and nonsubstituted phenyl-moleties. O-benzyl-p-chlorophenol, a hospital disinfectant that has widespread low level discharge to the environment through municipal wastewater treatment facilities, was rapidly biodegraded or photoxidized in river water and wastewater sludge samples, suggesting that its persistence in the environment was unlikely.[42] Biodegradation of o-phenylphenol, a chemical used widely in disinfectant formulations, in laboratory shake flask tests containing samples of either river water or activated sludge[43] was given as evidence to support the claim that this chemical would not persist in the environment. Similar conclusions were reached from biodegradation studies on ϵ-caprolactam.[44] This major precursor of nylon synthesis was used readily as a sole source of carbon and energy by microbial communities in lake water. The relationship between lipophilicity, substituent position on an aromatic ring and biodegradation was described by Paris *et al.*[45] Octanol-water partition coefficients, used as a measure of lipophilicity, were found to correlate with the biodegradation rate constants of carboxylic acid esters composed of a fixed aromatic ring moiety and increasing lengths of the alkyl component. Poor correlations were obtained with ethyl esters of chlorine-substituted carboxylic acids.

Microbial dehalogenation reactions continued to be investigated because of their importance in the fate of chlorinated pollutants. LaPat-Polasko *et al.*[46] studied a *Pseudomonas* species capable of using the substrates methylene chloride and sodium acetate simultaneously for carbon and energy, provided one was present at high concentrations (mg/L) and the other was present at low concentrations (μg/L). The authors explained that the faster utilization of trace concentrations of methylene chloride, a common environmental contaminant, in the presence of acetate than in its absence suggests that possible manipulation of substrate combinations may significantly enhance desirable biotransformations in the field. Sequential dehalogenation of trichloronitromethane (chloropicrin) to nitromethane by *Pseudomonas* isolates from soil was reported by Castro *et al.*[47] These reductive dehalogenations were apparently typical reactions of iron (II) porphyrins or heme proteins, such as the enzyme P-450 cam found in these pseudomonads. Klecka and Gonsoir[48] used *in vitro* experiments to verify that iron porphyrins, in the presence of a reducing agent, would catalyze the dehalogenation of several chlorinated methanes and ethanes. Because anoxic environments are likely to contain free iron porphyrins, chemical as well as biological processes may play an important role in determining the fate of chlorinated hydrocarbons in these evironments. Dechlorination of chlorobenzoates and chlorobenzenes was noted in most studies[13,49] to occur after ring cleavage. However, Marks *et al.*[49] convincingly verified again that dechlorination in aerobic environments might occur before ring cleavage. This mechanism seemed to be a halidohydrolysis reaction typical in the dehalogenation of other haloalkanoic acids. Cook and Hutter[69] characterized a similar hydrolytic dechlorination of deethylsimazine by bacteria. Wachter and Andelman[50] demonstrated that algal extracellular products can stimulate formation of

organohalides, including chloroform, during standard wastewater chlorination procedures. Kaczmar *et al.*[51] provided experimental data showing that the volatility of some of these organohalides would prevent their accumulation in aquatic environments. On the other hand, humic acids, as shown in experiments by Callaway *et al.*,[52] increased the solubility of low molecular weight halogenated hydrocarbons and, therefore, may have increased the persistence of these chemicals. Finally, in anticipation of future problems associated with the potential biodegradation of detergents and surfactants sorbed to surfaces, Urano *et al.*[53-54] provided sorption isotherms for two nonionic synthetic surfactants on sediments and microorganism surfaces.

PESTICIDES

Dobbs *et al.*[55] described a method for estimating vapor pressures of pesticides by examining loss rates of one pesticide relative to a standard pesticide in binary mixtures of the chemicals. Within a vapor pressure range of 10^4 to 0.2 Pa, and using a constant temperature oven with a controlled throughput of air, loss rates could be reproduced to within a coefficient of variation better than 11%. Vapor pressures were within a factor of 3 of theoretical. The major dissipative processes for the herbicide Drepamon in a rice field under typical field-use conditions was determined by Draper and Crosby[56] to be volatilization and photodecomposition, the former being the faster rate process. However, photochemical oxidants, such as trace quantities of hydrogen peroxide, may greatly enhance the photodecomposition rates. Photodegradation products from the oxime carbamates, aldicarb and methomyl, have been described by Freeman and McCarthy.[57-58]

Recent interest in pesticide-sediment-water interactions was reflected in several studies of adsorption-desorption kinetics and octanol-water partition coefficients.[59-60] Corwin and Farmer,[61] for example, indicated that adsorption and desorption of bromacil and diquat to freshwater sediments were not single-valued parameters (that is, an hysteretic effect) for a predetermined equilibrium time. Consequently, no single equation could describe both processes. Gerstl and Mingelgrin[62] have shown that correlations between equilibrium partitioning, solubility, and organic carbon were inconsistent between different soils and lake sediments. They attributed possible differences in humic acid chemical makeup, such as higher paraffinic hydrocarbon contents or organic matter-mineral relationships, as the source of these inconsistencies. Colloidal organic material in estuarine environments was shown[63] to enhance the solubility of atrazine by incorporation of the pesticide into stable complexes. Ionizable hydrophilic sites, such as amines, hydroxyl, phenolic, and carboxyl groups, on the resulting complex maintain its colloidal nature and significantly effect transport of the pesticide in aquatic environments. Kinetics and mechanisms for the chemical hydrolysis of aldicarb in groundwater[64-65] and chlorpyrifos in natural freshwater[66] were reported. Second-order rate expressions relating hydrolysis rate constants to pH were appropriate in all cases, except for the alkaline hydrolysis of chlorpyrifos.

Laboratory-stream microcosms were used to study the fate and effects of atrazine and hexachlorobiphenyl.[67] Actual distributions of the two chemicals could be adequately predicted, within a factor of 10, from information on mass transport, solubilities, and octanol-water partition coefficients. Microcosms containing environmental components from a local pond were also used to verify that atrazine was as persistent in complex systems as it was in simpler laboratory test systems.[68] Biodegradation of triazine herbicides, however, has been demonstrated in laboratory pure culture studies.[69,71] Cook and Hutter[69] isolated a bacterium capable of quantitative utilization of deethylsimazine, the first fungal breakdown product of a chloro-s-triazine, as a nitrogen source for growth. The degradation proceeded in two steps: a unique hydrolytic dechlorination followed by amination of a nitro group. Also described was a second type of dealkylation reaction of chlorinated s-triazines.[70] The marine fungus, *Periconia prolifica,* was shown by Schocken and Speedie[71] to simultaneously use two metabolic pathways to degrade atrazine, a hydrolysis to hydroxyatrazine and a dealkylation to deisopropylatrazine. The results suggested that the atrazine conversion to hydroxyatrazine in marine microcosms may be the result of fungal activities therein, a possibly important aspect of biodegradation not routinely considered.

The fate of methyl parathion in outdoor ponds and in laboratory microcosms containing different combinations of tap water, pond water, pond sediment, and rooted macrophytes was compared[72] and the importance of microbial communities associated with sediment and plant surfaces in the biodegradation of this pesticide was revealed. The utility of microcosms for characterizing biodegradation processes that occur in the field was emphasized, but caution in use of the generalized second-order rate approach for making predictions of biodegradation was recommended, unless accurate measures of the microbial biomass on these surfaces could be obtained. In microcosm systems studied by Holm *et al.*,[73] however, microbial activity associated with stream sediments did not affect the biotic fate of methyl parathion. Organic enrichments and Aufwuchs communities were the principal factors controlling the biodegradation of this pesticide. Fate studies on methyl parathion and linuron in small enclosures in farm ponds[74] also suggested that biodegradation is important in the fate of these pesticides. Linuron disappearance could be greatly enhanced by adding a bacterial culture known to biodegrade the pesticide to the enclosures. Badawy and El-Dib[75] showed that the biodegradation of methyl parathion in seawater involved an adaptation response, that is, respiking seawater with methyl parathion after complete disappearance of original methyl parathion resulted in a considerably shorter half-life for decay. Initial disappearance of methyl parathion resulted in the transient appearance of the reduction product, amino methyl parathion. After two subsequent respikes with the pesticides, dealkylation products, monomethyl parathion and monomethyl amino parathion, and the hydrolysis product, paranitrophenol, were detected. Distribution and movement of methyl parathion in water and sediment following a spill in the Mediterrean Sea was also described.[76]

In other pesticide degradation studies, the importance of living plant tissue on the fate of phorate in pond ecosystems and the effect of adjuvants in commercial preparations of pesticides on the fate of fenitrothion and aminocarb in model ecosystems was reported.[77-78] Caplan *et al.*[79] determined that fenvalerate in tidal marsh microcosms was metabolized, presumably by microorganisms in sediment, by either hydration of the cyanide functional group, hydroxylation of the diphenylether aromatic ring, or hydrolysis of the central ester linkage.

The herbicides 2,4.5-trichlorophenoxyacetic acid (2,4,5-T) and picloram were shown by Skurlatov *et al.*[80] to be more rapidly photolyzed in natural waters containing humic acid materials. The humic acid photosensitization may be the predominant factor affecting the fate of 2,4,5-T in natural aquatic systems because photolysis in the absence of the humic acid was very slow. Suflita *et al.*[81] noted that 2,4,5-T could be dechlorinated by an anaerobic methanogenic consortium of microorganisms growing on 3-chlorobenzoate. The biodegradation of 2,3,6-trichlorophenylacetic acid, a herbicide, in wastewater, lake water, and soil seemed to involve initial oxidation of the acetic acid sidechain and not hydroxylation of the aromatic ring.[82] However, ring oxidation and opening must eventually occur, at least in soils and wastewater, because mineralization of radiolabeled parent material to $^{14}CO_2$ occurred. Lewis *et al.*[83] attempted to use biodegradation rate information from laboratory flask studies to predict the biotic fate of 2,4 dichlorophenoxyacetic acid (2,4-D) in periphyton-dominated microcosms. Accurate predictions were possible if first-order decay rates were corrected for differences in the ratio of colonized surface area to container volume. Interestingly, because virtually all bacteria from the periphyton material could degrade the 2,4-D, differences found in total microbial biomass should have given different first-order rates. Absence of these differences, however, led the authors to suggest that where bacterial concentrations were low, difference in transformation activity was made up by increased activity of fungi or other microorganisms. Laboratory biodegradation studies of the herbicide fluridone (1-methyl-3-phenyl-5-[3-(trifluoromethyl)-phenyl]-4-pyridinone) using water-sediment systems, showed that one of the phenyl rings in the parent molecule was hydroxylated, opened and oxidized to a carboxyl functional group.[84] This product, however, was not detected in 20 different field ponds after the herbicide was applied; rapid aqueous photolysis so greatly predominated in natural sunlight that fluridone dissipated from the aquatic environment before the much slower microbial degradation occurred. Hydrolysis and sorption was shown, based on standard laboratory test protocols, to be an important processes controlling the fate of lindane in aquatic environments.[85] More rapid biodegradation of methoxychlor in anoxic sediments, as compared to water, was reported by Muir and Yarechewski.[86] They detected rapid formation of mono- and dihydroxy reductive dechlorination products at the appropriate redox potential in sediments, but noted no further decomposition of these products. A similar study with DDT, kepone, and permethrin showed that only DDT was significantly transformed under anaerobic conditions.[77]

POLYCYCLIC AROMATIC HYDROCARBONS

Sorption properties of polycyclic aromatic hydrocarbons (PAHs) have been characterized in terms of partitioning from water into octanol,[88] suspended solids in coal-coking wastewater,[89] and activated carbon.[90] Wijayaratne and Means[91] examined the sorptive behavior of natural estuarine colloids, with benzene, naphthalene, and anthracene, and found that Freundlich partition coefficients normalized to organic content of the colloids were an order of magnitude higher than corresponding soil/sediment systems. The higher affinities exhibited by the natural colloids were discussed in terms of possible greater positional availability in sites for hydrophobic binding and charge density separation. PAH concentrations in Washington coastal sediments were shown by Prahl *et al.*[92] to originate largely from suspended particulate material discharged from the Columbia River (30%) and to a lesser extent (10%) from atmospheric inputs.

Biodegradation of PHAs by bacteria and fungi was reviewed extensively by Cerniglia.[93] Oxidation of the methyl group of 1- and 2-methylnaphthalene was reported by Cerniglia *et al.*[94] to be the initial metabolic transformation catalyzed by the fungus, *Cunninghamella elegans*. This activity represents a potential detoxification pathway for the methyl naphthalene in nature. The bacterial oxidation of acenaphthene and acenaphthylene to their respective quinones was studied by Schocken and Gibson.[95] The authors used a species of *Beijerinckia* to show that different pathways were involved in converting the parent material to acenaphthene quinone. Two successive monooxygenations and two dehydrogenations were involved in acenaphthene oxidation, whereas acenaphthylene oxidation seemed to proceed through an initial dioxygenation step followed by the dehydrogenation steps. Boyle *et al.*[96] reported that fluorene disappearance from experimental pond ecosystems was controlled primarily by evaporation. Evidence for evaporation was based on agreement between the observed half-life of fluorene in the pond and the half-life calculated from an algorithm of vapor pressure. Rate of fluorene disappearance significantly correlated with algal productivity and dissolved oxygen concentrations at dawn. The condition led the authors to conclude that degradation, by either photolysis or microbial activities, may also have been occurring in the ponds. Photolytic decay to a quinone and, to a lesser extent, adsorption to sediments were the principal processes affecting the fate of anthracene in a shallow clear water artifical stream channel.[97] Biodegradation, which was expected based on laboratory studies, to be an important fate process, did not occur in the stream channel. Model simulations that reasonably described the fate of anthracene in the channel were noted by Bartell *et al.*[98] to be possible, with the use of structure-activity relationships. Error analysis also revealed that the precision of predicted concentrations depended on precise estimations of model parameters (water solubility, photolytic decay, volatilization, sorption, and bioaccumulation), and greater precision was obtained from predictions of relative distribution in the ecosystem components than from predictions of concentration within each component.

HEAVY METALS

Cadmium and copper distributions between estuarine sedimentary phases and water were satisfactorily predicted with a simple model based on speciation among soluble ligands.[99] Cadmium-binding to sediments was determined to be controlled primarily by iron oxides and to a lesser extent by humic acid organics, whereas copper-binding was determined predominately by the humic acids. Maher[100] described a sequential extraction scheme to study the associations of trace metals to estuarine and oceanic sediments. Santschi *et al.*,[101] used historical analysis of heavy metal distributions in sediment cores from Narragansett Bay to conclude that with a 20 to 30% uncertainty, most of particle-reactive radionuclides (^{210}Pb and $^{239,240}Pu$) and trace metals entering the bay are efficiently

transferred to the sediments and retained within the bay system. In addition, 7×10^7 kg (69 000 tons) of sediments, 5.2×10^4 to 9.1×10^4 kg (51 to 90 tons) of Pb, 7.3×10^4 to 10.2×10^4 kg (72 to 100 tons) of Cu and 4×10^5 to 10.2×10^5 (400 to 1000 tons) of hydrocarbons per year now accumulate in the bay. The distribution of metals in the easily exchangeable, oxidizable, reducible, and resistant fractions of estuarine sediments was determined by a sequential leaching technique.[102] All metals, especially iron, were predominately associated with the resistant fraction, although association with the oxidizable organic fraction was also frequent. Renwich and Edenborn[103] have showed detected high correlations among sediment texture, total coliform bacteria, and metal concentrations in New Jersey estuarine sediments which, as a result, may allow a reduction in the number of field and laboratory tests required before dredging or other site-modifying activities. They further reported that heavy metal concentrations were high, not only near industrial sites, but also near urban areas, where surface runoff and contamination from pleasure craft were present. Talbot and Chegwidden,[104] however, concluded that sediments were not a good indicator of metal contamination in estuarine areas influenced by industrial activities because of higher concentrations in water and biota. Further studies by Talbot[105] noted the importance of speciation in controlling the fate of heavy metals in aquatic ecosystems. Heavy metals entering a harbor near a wharf area bound quickly to settling particulate material, causing elevated concentrations in surface sediments, whereas metals entering a flats area accumulated in filter-feeding mollusks, rather than sediment. Copper and zinc, entering an estuary in cooling tower water, were shown by Waslenchuk[106] to readily partition between solid and liquid phases during mixing, so that the effects of particulate-metals on local biota were greatly reduced. Paulson *et al.*[107] also demonstrated that the chemical forms of trace metals, rather than their sources, were important in the physicochemical reactions affecting the fate and distribution of iron, manganese, copper, and cadmium in an estuary. Associations and distributions of metals in sediments were characterized in other studies.[108–110] Attempts to develop pollution indexes of metals in sediments have been addressed statistically by Nishida and Suzuki,[111] and Nishida *et al.*[112] The effect of microbial biomass on the removal of metals from activated sludge wastewater was found to be related primarily to the physical characteristics of the particulates, rather than their viability.[113]

Photolysis of tributyltin in natural waters by sunlight was described as a slow ($t_{1/2} > 89$ days), but significant environmental process in the fate of this organometal, as other fate processes are slower.[114] Direct photolysis seemed to involve, possibly through a free radical mechanism, sequential debutylation to inorganic tin. Correspondingly, inorganic tin was detected in sediments contaminated by tributyltin.[115] However, detectable concentrations of parent material, as well as methylated forms of tin, tri-n-butylmethyltin and di-n-butyldimethyltin, suggest that degradation processes are slow and possibly catalyzed by microorganisms. Craig and Moreton[116] established that sulfide mediated the decomposition of methylmercury, probably through a dismutation, by bis-methylmercury sulfide to dimethylmercury. The dimethylmercury then volatized out of contaminated sediments. The synthesis and stability of monomethylmercury in estuarine sediments was noted by Compeau and Bartha[117] to be enhanced by anaerobic, low salinity conditions. Their experimental evidence suggested that low salinity was associated with sulfate reduction to sulfide, a product which would form a precipitate with Hg^0. The anaerobic conditions would promote either reduction of Hg^{2+} to Hg^0 or disproportionation by H_2S to volatile dimethylmercury.

CRUDE OIL AND HYDROCARBONS

Physical factors affecting the dissolution of petroleum hydrocarbons in aquatic media were studied in conjunction with volatilization processes,[118,119] tar ball formation, distribution,[120,121] and leaching into aqueous phases[122] and sedimentation of oil-contaminated particulates (detritus and plankton) in estuarine areas,[123–124] microcosms,[125] and artifical enclosures.[126] Stiver and Mackay[119] developed a theoretical framework, incorporating effects caused by surface to volume ratio, wind speed and time of exposure, for quantifying evaporation rates of crude oils. Gas-stripping and tray-evaporation techniques proved most ideal for measuring evaporation rates of crude oils in the laboratory and for producing weathered samples of crude oil. Weathering of artificial tar balls, prepared in the laboratory from four different crude petroleums,[119] resulted in a decrease of aromatic hydrocarbons and alkanes, but an increase in nitrogen-, sulfur-, and oxygen-containing hydrocarbons. Composition and weathering of the artificial tar balls were similar to that reported for natural tar balls. Chemical analysis of water-soluble fractions of oils showed a predominance (greater than 90% of the water-soluble fraction) of aromatic hydrocarbons, particularly alkylbenzenes, alkylnaphthalenes and xylenes.[121–122] The water-soluble fraction was affected largely by the composition of the oil and temperature (lower solubility at lower temperature), rather than by turbulent mixing within water column. Tjessem *et al.*[127] demonstrated that the addition of a detergent, Corexit 9527, to an oil slick contained in a large plastic enclosure of seawater caused the oil to remain at the water surface, the most reactive part of the water column. The accumulation of large amounts of polar constituents suggested that the detergent promoted photochemical transformations of the hydrocarbons. Photolytic transformation products were observed in another study of oil weathering.[128] However, elimination of the contained planktonic population suggested these products may be toxic. Kuiper *et al.*[129] reported that bioturbation activities in sediments were responsible for the rapid burial of oil added to tidal flat microcosms. Although oil mousse on the sediment surface eventually disappeared, sediment-bound hydrocarbons were continuously released into the water column of the microcosm, causing depressions in primary production of plankton in the water and stimulations in phytobenthos biomass. Decreased grazing activities and high mortalities of macrofauna were also caused by this prolonged exposure to the oil.

The importance of biodegradation in the fate of oil and specific classes of petroleum hydrocarbons (saturates, aromatics, asphaltenes) was assessed by Fusey and Oudot,[130] by chemical and graphical methods to interpret the weathering of Arabian light crude oil spread on the surface of seashore sediments. This method permits the relative parts of physical and biological removal processes involved in weathering to be estimated. Biodegradation of aromatic hydrocarbon components in crude

oil added to sediments in the field was verified by showing that the unresolved mixture of hydrocarbons, or hump (as seen in chromatographic analysis), remaining after biodegradation of the aromatic fraction in laboratory experiments was also observed in the field studies.[131] London *et al.*[132] demonstrated that the water-soluble fraction, primarily aromatic hydrocarbons, of petroleum- and shale-derived fuels will support the growth of microorganisms in the absence of other carbon sources. The authors suggested the unresolved complex mixture could be used as a marker to recognize past contamination of sediment with crude oil. The biodegradation of sulfur and nitrogen heterocyclic hydrocarbons in the aromatic fraction of crude oil was studied by Fedorak and Westlake.[133–134] Mixed microbial cultures from soil enrichments readily degraded both chemical types; the fewer the number of alkyl carbons on both the carbazole and benzothiophene rings, the more susceptible the hydrocarbons were to microbial attack. Pure cultures of bacteria isolated from tanker ballast water will also degrade significant amounts of light crude oil.[135] Characterization of their ability specifically to degrade alkylnaphthalenes revealed a dependence of the degradation on the position, number, and type of alkyl substituents. Meyer *et al.*[136] provided significant evidence that diauxic growth relationships in mixed cultures of bacteria alter degradation patterns of hydrocarbons in unpredictable ways. Phenol, for example, was reported to inhibit the degradation of benzene and naphthalene; even the mixed culture would metabolize all hydrocarbons singly. Finally, Hou *et al.*[137] further characterize the biodegradation, through initial monoxygenase-catalyzed epoxidation, of short-chain alkenes by pure cultures of propane-using organisms.

FATE PROCESS CHARACTERIZATION

To assess and predict the fate of pollutants in aquatic environments depends, in part, on knowledge of the theoretical and mechanistic details of individual fate processes without particular reference to a specific chemicals or chemicals. In this context, definitive studies of hydrolytic processes,[138] photolytic reactions,[139–140] volatilization,[141] solubility,[142] and sorption mechanisms[143–145] should be mentioned as potential sources of detailed information on these fate processes. A unique study by Karickhoff and Morris[146] reported quantitatively the influence of tubificid oligochaetes on the movement and distribution of pollutants in sediments. The time required for the mixing of hydrophobic pollutants was independent of the physical properties of the chemicals and was controlled by physical movement of sediment particles by the bioturbation activities of the invertebrates.

With respect to biodegradation processes, the effects of fulvic acid,[147] exudates from algae, fungi and bacterial communities,[148] and microbial biomass[149] were studied. Lewis *et al.*[150] and Simkins and Alexander[151] have examined, both theoretically and experimentally, the kinetics of biodegradation processes. Numerous studies on the methodology, interpretation, and meaning of standardized biodegradability testing were reported.[152–157] Finally, Pritchard and Bourquin[158] comprehensively reviewed the use of microcosms as a tool for evaluation of interactions between pollutants and microorganisms. Perez[159] has discussed similar uses of microcosms, but more in the light of their application to standardized testing protocols for ecological fate and effects assessment.

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